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Cluster Formation and Evolution on Semiconductor and Insulator Surfaces

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Final Report

Part I

Papers Published in Refereed Journals under ONR N00014-87-K-0029

1. Yoram Shapira, F. Boscherini, C. Capasso, F. Xu, D.M. Hill, and J.H. Weaver, "Au/InSb(110) Interface Profiles from Synchrotron Radiation and Polar-Angle-Dependent X-Ray Photoemision," Phys. Rev. B **36**, 7656-7659 (1987).

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1. Y. Gao, T.J. Wagener, D.M. Hill, H.M. Meyer III, J.H. Weaver, A.J. Arko, B. Flandermeyer, and D.W. Capone II, "High Temperature Superconductors: Occupied and Unoccupied Electronic States, Surface Stability, and Interface Formation," Chapter 21 in *Chemistry of High-Temperature Superconductors*, David L. Nelson, M. Stanley Whittingham, and Thomas F. George, eds. (American Chemical Society, Washington, D.C., 1987) ACS Symposium Series 351, pp. 212-225.

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1. G.D. Waddill, I.M. Vitomirov, C.M. Aldao, S.G. Anderson, C. Capasso, and J.H. Weaver, "Metal-Semiconductor Interfaces with Novel Structural and Electronic Properties: Metal Cluster Deposition," in *Chemistry and Defects in Semiconductor Heterostructures*, Proceedings of the 1989 Spring Meeting of the Materials Research Society, Vol. 148, edited by M. Kawabe, E.R. Weber, T.R. Sands, and R.S. Williams.

Patents Filed

J.H. Weaver, H.M. Meyer III, D.M. Hill, D.L. Nelson, and R.K. Grasselli, "Ohmic Contacts and Passivation Layers for High Temperature Superconductors."

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Invited Presentations at Topical or Scientific/Technical Society Conferences

1. J.H. Weaver, Panelist in Workshop on Microelectronics Research and Development, Congress of the United States, Office of Technology Assessment, Washington, DC, June 1987.

2. J.H. Weaver, "Growth and Characterization of Metal Overlayers on Compound Semiconductors," American Institute of Chemical Engineers, Conference on Emerging Technologies in Materials, Minneapolis, Minnesota, August 1987.

3. J.H. Weaver, "High Temperature Superconductors: Occupied and Unoccupied Electronic States, Surface Stability, and Interface Formation," American Chemical Society, New Orleans, Louisiana, September 1987.

4. J.H. Weaver, H.M. Meyer III, Y. Gao, D.M. Hill, T.J. Wagener, B. Flandermeyer, and D.W. Capone II, "High Temperature Superconductors: Occupied and Unoccupied Electronic States," 20th Annual Synchrotron Radiation Center Users Group Meeting, Stoughton, Wisconsin, October 1987.

5. Zhangda Lin and J.H. Weaver, "Surface Segregation at Metal/III-V Semiconductor Interfaces," Solvay Conference on Surface Science, Austin, Texas, December 1987.

6. J.H. Weaver, "High Temperature Superconductors," American Institute of Chemical Engineers Symposium, Minneapolis, Minnesota, February 1988.

7. J.H. Weaver, "Surface Stability and Interface Chemisorption of High Temperature Superconductors," Gordon Conference on the Chemistry of Electronic Materials, Ventura, California, March 1988.

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9. J.H. Weaver, "The Physics and Chemistry of Interfaces," keynote lecture, The Royal Society of Canada, Symposium on Advanced Materials: Science & Applications, Windsor, Ontario, Canada, June 1988.

10. J.H. Weaver, "Electronic Structure of High Temperature Superconductors," Gordon Conference on Electron Spectroscopy, Brewster Academy, Wolfeboro, New Hampshire, July 1988.

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12. J.H. Weaver, "Metal-Semiconductor Interfaces," World Materials Congress: Interface Science and Engineering, Chicago, Illinois, September 1988.

13. J.H. Weaver, "High Temperature Superconductors: Electronic States, Surface Stability, and Interface Formation," 35th National Symposium of the American Vacuum Society, Topical Conference on High T_c Superconducting Thin Films, Devices, and Characterization, Atlanta, Georgia, October 1988.

14. J.H. Weaver, "Physics and Chemistry of Metal/Semiconductor Interfaces," Spring Meeting of the Materials Research Society, Symposium on the Chemistry and Defects in Semiconductor Heterostructures, San Diego, California, April 1989.

15. J.H. Weaver, "Surfaces and Interfaces of High Temperature Superconductors," First Gordon Conference on Superconductivity, Ventura, February 1989.

16. J.H. Weaver, "Photoemission Studies of Surfaces and Interfaces of High T_c Superconductors," Scanning Electron Microscopy Conference, Salt Lake City, Utah, May 1989.

17. J.H. Weaver, "Physics and Chemistry of Metal/Semiconductor Interfaces," American Vacuum Society, Albuquerque, May 1989.

18. J.H. Weaver, "Reactions at Semiconductor Interfaces," Special Lecture Series on Electronic Materials: A New Era of Materials Science, University of Minnesota, Minnepolis, May 1989.

Contributed Presentations at Topical or Scientific/Technical Society Conferences

1. J.J. Joyce, C. Aldao, B.M. Trafas, and J.H. Weaver, "Island Formation and Metallic Screening at the In/Semiconductor Interface," 20th Annual Synchrotron Radiation Center Users Group Meeting, Stoughton, Wisconsin, October 1987.

2. I.M. Vitomirov, F. Xu, C.M. Aldao, and J.H. Weaver, "Direct Evidence of the Onset of In Surface Segregation for Co/InP(110)," 34th National Symposium of the American Vacuum Soceity, Anaheim, California, November 1987.

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6. J.H. Weaver, "Physics and Chemistry of Interfaces," University of Washington, Seattle, May 1988.

7. J.H. Weaver, "Physics and Chemistry of Interfaces," Battelle Pacific Northwest Laboratory, Richland, Washington, May 1988.

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21. J.H. Weaver, H.M. Meyer III, D.M. Hill, and T.J. Wagener, "Surfaces and Interfaces of High T_c Superconductors," Physics Department, Texas A&M University, College Station, September 1989.

Honors/Awards/Prizes

None

Personnel Supported under ONR N00014-87-K-0029 (Full or Partial Support)

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Summary of Accomplishments under ONR N00014-87-K-0029

The work performed under this program can be divided into three main groups. The first involved the formation of metal–semiconductor interfaces by atom deposition, with emphasis on the development of clusters on the surface, the onset of disruption, the nucleation of the metal, and the kinetics of atom redistribution in the evolving overlayer. The second involved the surface and interface properties of the high temperature superconductors. The third involved the formation of abrupt interfaces using a novel technique that we developed, namely cluster-assembly. These programs were very productive, as summarized above. Fifty-three refereed papers and seven chapters were published, eighteen invited papers were given, twenty-one contributed papers were presented, one patent was filed, and eleven students and postdocs received partial support. Major progress was possible because of the momentum developed through ONR support and state-of-the-art instrumentation that had been developed in our laboratory, largely through ONR support. Contributing to this ONR-Chemistry program was support from ONR/DARPA for HTS studies late in this contract period and support from SDIO under a program for reliable electronics.

Common to all of our studies is the need to understand the detailed chemistry of a surface as it evolves from its freshly-prepared state to its final state where an overlayer has grown over the surface Overlayer growth is accomplished by atom deposition and cluster-assembly, and we have shown that quite different final configurations can be reached because the reaction pathways are fundamentally different.

Our studies of atom-assembly of metal overlayers on semiconductors pioneered the use of high resolution synchrotron radiation photoemission to investigate interface reactions at low coverage. Primary studies examined the growth of epitaxial and nonepitaxial metal layers on GaAs, InSb, and InP. We completed detailed investigations of the evolution of the Schottky barrier as a function of temperature and dopant concentration, showing the importance of nonequilibrium processes associated with the surface photovoltaic effect and having a major impact on the understanding of surface processes. We showed that atom-assembled interfaces are rarely atomically abrupt, even in instances where reaction is weak. Almost all cases investigated showed intermixing, the degree of which varied according to the chemical reactivities of the constituents. Temperature-dependent studies of representative systems, including Ti/GaAs, Cr/GaAs, Ag/GaAs, and Bi/GaAs demonstrated the role of thermal energy and kinetics on the initial stages of reaction and on subsequent atom distributions in thickening films. The mechanisms behind surface segregation were examined in detail, and direct evidence was given for the onset of segregation phenomena.

Our studies of surface and interface properties of the high temperature superconductors broke new ground and provided broadly-based fundamental understanding of surface stabilities and materials compatibility issues for the HTSs. The importance of such insight is evident from the need to integrate the HTSs with new and existing technologies. By initiating our interface studies immediately after the discovery of the HTSs, we established ourselves as leaders in the field. Our work focused on the surface chemistry for all of the HTS materials developed to date, namely the 2-1-4, 1-2-3, 2-2-1-2, and 2-2-2-3 materials, with various elemental substitutions and ranges in stoichiometry. These HTS materials are based on Cu-O structures and, as such, they exhibit similar, but not identifical, surface properties. (The exception is BaKBiO, which we also studied) Our results showed that the HTS surfaces are fragile, being readily degraded by Ar ion bombardment, exposure to environmental gases, and the deposition of almost all atoms. Interface phenomena for these systems are complex, but not intractibly so.

In our surface and interface studies of HTSs, we established collaborations with those who were expert in the synthesis of bulk samples. This made it possible for us to change directions rapidly as new superconductors were developed. It also allowed us to provide important characterization of the materials that have become available. These collaborations involved colleagues from Argonne, Los Alamos, Sandia, 3M, Grace, Honeywell, ONR, and most recently, colleagues from Japan and China.

Under this program, we examined a wide variety of reacting overlayers, including transition metals (Ti, Fe), noble and near noble metals (Cu, Pd), rare earths (La), simple metals (Al, In), and semiconductors (Ge, Si). These interfaces exhibited oxygen removal from the HTS, the loss of superconductivity in the modified region, the formation of oxides, and, ultimately, the appearance of metal overlayers. These interfaces were also metastable. All led to non-ohmic and heterogeneous boundary layers between the metal and the buried superconductor. In contrast, we showed that Ag and Au overlayers were not disruptive and that Bi overlayers were minimally reactive, indicating the significance of these materials as contacts. We examined the feasibility of preparing passivating surface layers, with emphasis on Bi and Bi_2O_3, the oxides of Al and Si (formed in activated oxygen atmospheres), and CaF_2. These latter studies showed great promise for passivating dielectric layers, and patent application procedures were initiated.

The third major thrust of our work involved development of a new technique for the assembly of interfaces, namely cluster-assembly. Previous work had involved atom-by-atom deposition and overlayer growth, and we showed the importance of surface chemistry in understanding the details of film nucleation and growth. For cluster-assembly, our goal was to form an interface in a way that changed the surface chemistry by preventing single atoms from contacting the surface. The technique involves the formation of a thin buffer layer on a surface such that it serves as a noninteracting support for subsequent depositions. When metal or semiconductor atoms are deposited onto the buffer layer, they nucleate and grow as clusters. The contact of these clusters with the surface is accomplished by desorbing the Xe. The result is an atomically abrupt interface in most cases. Development of this technique generated a great deal of interest because of the opportunity to form novel junctions. For the HTS materials, for example, we succeeded in growing metal overlayers without disruption of the HTS surface. For semiconductor contacts, we found novel Fermi level pinning positions. On-going studies focus on the properties of these interfaces, issues related to sintering of nanoclusters and the boundary layer itself, and the possibility for novel device structures.